

Nanostructure fabrication of zero-dimensional quantum dot diodes

J. N. Randall, M. A. Reed, R. J. Matyi, and T. M. Moore

Central Research Laboratories, Texas Instruments Inc., Dallas, Texas 75265

(Received 3 June 1988; accepted 2 August 1988)

The nanofabrication techniques which are used to create quantum dot diodes will be discussed. The device is a vertical resonant tunneling diode where the lateral dimensions are reduced to ~ 1000 Å. Electron beam lithography is used to pattern a small self-aligned metal dot top contact and etch mask. The semiconductor dot is a cylinder ~ 1000 Å in diameter and on the order of 100 Å thick, with the vertical potential defined by the double-barrier heterostructure and the lateral defined by the Fermi-level pinning of the free surfaces. Discrete energy states due to the three-dimensional confinement are observed and are spin degenerate only. Transport measurements through such a device will be presented.

I. INTRODUCTION

Ultrasmall electronic devices operating on quantum-mechanical effects are interesting both to the physics and engineering communities. Quantum size effects in two-dimensional, one-dimensional, or zero-dimensional structures create energy bands or discrete states which do not exist in larger devices. Experiments with such structures have advanced our understanding of quantum physics and suggest the possibility of an integrated circuit technology with an extremely large functional density.¹ In this paper we will describe the nanofabrication techniques used to produce electronic devices which include zero-dimensional structures (quantum dots). The approach used to produce quantum dot nanostructures suitable for electronic transport studies was to laterally confine resonant tunneling heterostructures with a fabrication-imposed potential.² This approach embeds a quasibound quantum dot (or dots) between two quantum wire contacts. We will present transport measurements made with these devices.

II. QUANTUM DOT DIODES

We have fabricated quantum dot diodes which are based on resonant tunneling diodes (RTD's). In RTD devices a thin layer (a quantum well) of semiconducting material is clad by two layers of a higher-band-gap material which are thin enough to allow tunneling.³ The current peak in the I - V characteristics of a RTD occurs under bias conditions where conduction-band electrons outside the barriers are energetically aligned with the quasibound electron states in the well, allowing tunneling through those states. In all cases reported here the RTD structure has been grown on GaAs using AlGaAs barriers and GaAs or InGaAs quantum wells.

The electron states in the well of a RTD are not discrete but are a parabolic band of states arising from the confinement of the electrons in one dimension. For electrons with momentum only in the direction normal to the plane of the quantum well, the band appears to have a discrete energy level.

Because the confinement structure is created epitaxially, the current flow is vertical and the lithographically defined dimensions of a RTD do not need to be particularly small. The quantum dot diodes described in this paper, on the other

hand, are RTD's with lateral dimensions small enough to create quantum size effects due to the lateral confinement. This additional confinement should split the parabolic band of confined electrons into discrete states.

In addition to the conventional double-barrier RTD, we have also created quantum dot diodes from epitaxial structures which included three barrier layers and two quantum wells. The widths of the wells were designed so that confined states in the two different wells and the conduction-band electrons would simultaneously come into alignment under one direction of bias. A laterally quantized version of this structure thus contains two quantum dots and permits the investigation of transport between the discrete states in the dots. To distinguish between the two different types of quantum dot diodes in this paper, we will henceforth refer to them as single-dot diodes and double-dot diodes.

III. FABRICATION PROCESS

The epitaxial structure of the single-dot diode consists of a $0.5\text{-}\mu\text{m}$ n^+ GaAs contact (Si-doped at $2 \times 10^{18} \text{ cm}^{-3}$, graded to $\sim 1 \times 10^{16} \text{ cm}^{-3}$ over 200 Å, followed by a 100-Å undoped GaAs spacer layer), a 40-Å $\text{Al}_{0.25}\text{Ga}_{0.75}\text{As}$ tunnel barrier, and a 50-Å undoped $\text{In}_x\text{Ga}_{1-x}\text{As}$ quantum well (x values studied ranged from 0 to 0.08). The structure was grown to be nominally symmetric about a plane through the middle of the quantum well. Large-area ($2\text{-}\mu\text{m}$ -square) mesas of a typical structure ($x = 0.08$) fabricated by conventional means exhibited two resonant peaks: a ground state at 50 mV with a peak current density of 30 A/cm^2 , and an excited state at 700 mV with a peak current density of $8.1 \times 10^3 \text{ A/cm}^2$, both measured at 77 K.

The epitaxial structure of the double-dot diode consists of a $0.5\text{-}\mu\text{m}$ n^+ GaAs contact (Si-doped at $2 \times 10^{18} \text{ cm}^{-3}$, graded to $\sim 1 \times 10^{16} \text{ cm}^{-3}$ over 200 Å, followed by a 150-Å undoped GaAs spacer layer), a 60-Å $\text{Al}_{0.4}\text{Ga}_{0.6}\text{As}$ tunnel barrier, a 80-Å undoped GaAs quantum well, a 35-Å interwell $\text{Al}_{0.4}\text{Ga}_{0.6}\text{As}$ tunnel barrier, a 50-Å GaAs quantum well, a 60-Å $\text{Al}_{0.4}\text{Ga}_{0.6}\text{As}$ tunnel barrier, and a top contact nominally identical to the bottom contact (except reflected through the z axis). The resonances are designed to be coincident for electron injection into the 80-Å quantum well first. Large-area ($2\text{-}\mu\text{m}$ -square) mesas of a typical structure

fabricated by conventional means exhibit a coincident resonant peak with a current density of 10 A/cm^2 . The coincident resonance is theoretically expected at 168 mV (for large-area mesas) for injection into the 80-Å quantum well. This is experimentally measured to be 170 mV.

The nanofabrication procedures used subsequent to the molecular-beam epitaxial (MBE) growth were essentially the same for both devices and the basic techniques have been reported previously.² A schematic of a completed quantum dot diode is shown in Fig. 1. We will summarize the process.

High-resolution electron beam lithography was used to define dots with diameters ranging from 1000 to 2500 Å. Lift-off was used to transfer the pattern to Ohmic metal dots. The evaporated metal layers which formed the dots were, in order of evaporation, 50 Å Ni, 25 Å Ge, 350 Å Au, 140 Å Ge, 115 Å Ni, and 500 Å Au. These dots must serve the dual purpose of etch mask and Ohmic contact. Reactive ion etching (RIE) was used to etch the dot pattern down through the RTD structure producing quantum dots. The RIE process used BCl_3 and was designed to produce highly anisotropic etching while maintaining a reasonably high GaAs-to-Au etch ratio. Next the structure was planarized with a polyimide layer which was etched back with O_2 RIE to uncover the top of the devices. Finally, a gold metal layer was patterned which connected the quantum dot diodes to bonding pads. A backside Ohmic metal layer was used as the other terminal.

IV. TRANSPORT

Before presenting transport measurements for single- and double-dot diodes, it is constructive to consider the effect of Fermi-level pinning at 0.7 V below the conduction-band edge for the free GaAs surfaces. The result is a depletion region which has a depth that is a function of the doping level in the GaAs. In the case of the smallest devices, this depletion region can significantly reduce the volume of device which is conducting. In fact, if the diameter of the device is too small, the entire device is depleted and no conduction is observed.

Another effect of the Fermi-level pinning is that the potential boundaries of our quantum dots are a slowly rising

radial function in contrast to the abrupt heterostructure interfaces at the barrier layers. Figure 2 schematically illustrates the lateral (radial) potential of a column containing a quantum dot, and the spectrum of three-dimensionally confined electron states under zero and applied bias. A spectrum of discrete states will give rise to a series of resonances in transmitted current as each state drops below the conduction-band edge of the injection contact. To observe lateral quantization of quantum well state(s), the physical size of the structure must be sufficiently small that quantization of the lateral momenta produces energy splittings $> kT$. Concurrently, the lateral dimensions of the structure must be large enough such that pinch-off of the column by the depletion layers formed on the sidewalls of the GaAs column does not occur. Due to the Fermi-level pinning of the exposed GaAs surface, the conduction band bends upward (with respect to the Fermi level), and where it intersects the Fermi level determines, in real space, the edge of the central conduction path core. When the lateral diameter is reduced to twice the depletion depth or less, the lateral potential becomes parabolic though conduction through the central conduction path core is pinched off. A structure that satisfies both constraints was achieved using a $\text{In}_{0.8}\text{Ga}_{0.2}\text{As}$ quantum well double-barrier structure with a physical (lithographic) lateral dimension of 1000 Å.

Transport measurements through the single-dot diode have been previously reported.⁴ Figure 3 shows the current-voltage characteristics of this (single) microstructure at 1.0 K. This fine structure in the I - V curve is evident at temperatures as high as 60 K. Assuming that the current density through the structure is approximately the same as a large-area device, measurement of the peak resonant current implies a minimum (circular) conduction path core of 130 Å for this structure; thus, a lateral parabolic potential approximation seems reasonable. This implies a depletion depth W of 430 Å at the double-barrier structure, in reasonable agreement with that expected from the known doping level (at $2 \times 10^{18} \text{ cm}^{-3}$, $W = 220 \text{ Å}$) and realizing that W will enlarge in the undoped double-barrier region. Assuming an approximate parabolic confining potential and a potential height of 0.7 eV, the discrete electron levels should be split evenly by 26 meV.

Only the excited-state resonance of this structure is observed since the current expected from the ground-state resonance is at the detection limits of the test apparatus. There appears a series of peaks appearing at approximately the same bias where a single negative differential resistance peak is expected in large-area diodes. In the range 0.75–0.9 V, the peaks are approximately equally spaced, with a splitting of $\sim 50 \text{ mV}$. Assuming that most of the bias is incrementally dropped across the double-barrier structure, the splitting of the equally spaced series is 25 meV, in excellent agreement with the value determined from the physical dimension of the structure. We believe that the structure observed here corresponds to resonant tunneling through the spectra of discrete quasibound (in the z direction) states in the quantum dot which correspond to the density of states of a three-dimensional semiconductor quantum well. Another peak, presumably the ground state occurs 80 mV below the equally

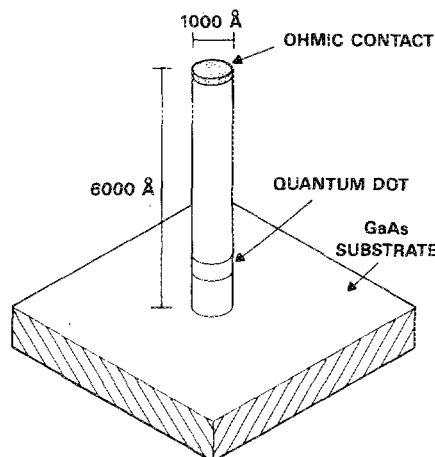


FIG. 1. Schematic of quantum dot diode structure.

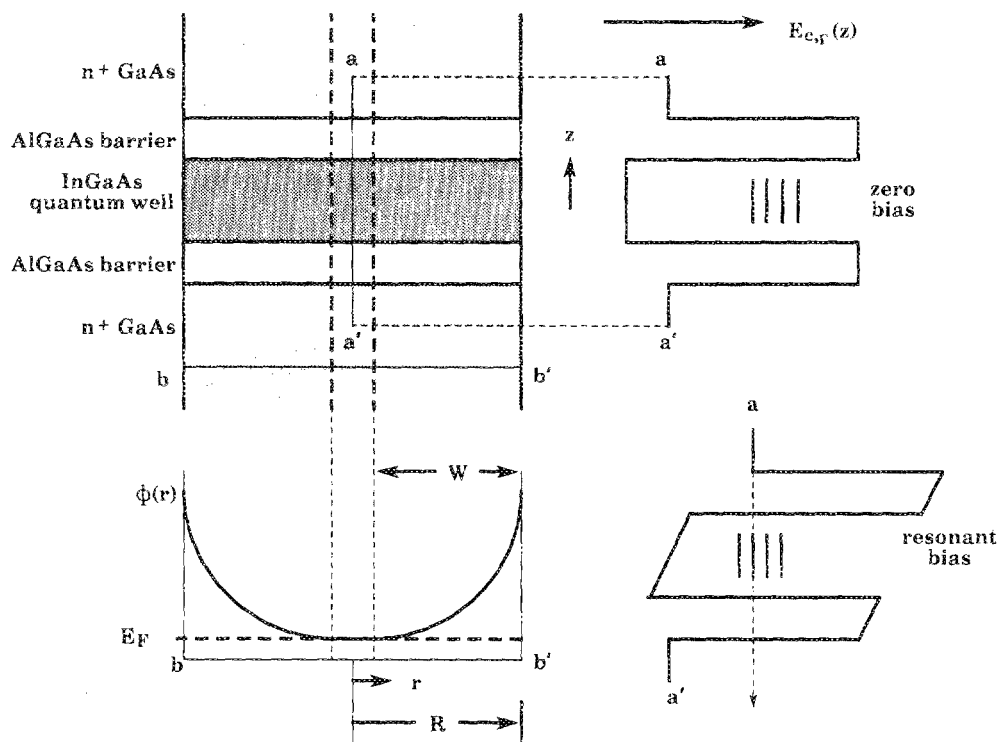


FIG. 2. Schematic illustration of the vertical (a-a') and lateral (b-b') potential of a column containing a quantum dot, under zero and applied bias. $\Phi(r)$ is the (radial) potential, R is the physical radius of the column, r is the radial coordinate, W is the the Fermi-level (E_F) pinning, and $E_{c,\Gamma}$ is the gamma-point conduction-band energy.

spaced series. The origin of this anomalously large splitting cannot be understood in terms of our simple harmonic oscillator potential. Efforts are underway to obtain a more realistic potential for this device based upon solving Poisson's equation self-consistently with the distribution of dopants and carriers, band-structure discontinuities, and the applied bias.

It has been suggested that tunneling through discrete states in a laterally quantized structure would not be observable if the symmetry of the structure were to permit separation of transverse and longitudinal variables in the Schrodinger equations.⁵ However, there exist a number of mechanisms that can break this symmetry; different effective masses in the heterostructure, lateral potential variations, scattering by nonsymmetrically placed impurity

atoms, etc. The observation here of discrete electronic states indicates that such symmetry is broken (i.e., "mode mixing").

A very intriguing situation exists for transport through the double-dot structures, since one can examine the tunneling between coupled discrete states. Figure 4(a) shows the I - V characteristics of one of these devices at 100 K. The lateral size of this device is $\sim 0.25 \mu\text{m}$. Two peaks are observable; the splitting of the peaks is 12 meV (contact resistance has shifted the peaks from 170 mV to ~ 1.7 V). These are the two quantum well resonances, split from coincidence. We have not yet determined the mechanism for the splitting (~ 10 meV); possibilities are fabrication-imposed strain or quantum well fluctuations.

Figure 4(b) then shows the same structure cooled to 4.2 K, and exhibits the various phenomena observed in these structures. First, the "noise" at 1.4–1.5 V is not external noise, but "telegraph" switching due to single electron trapping; likewise, the large "jump" at 1.9 V is also due to this effect.⁶ Second, a series of small peaks surrounding the major peaks has appeared. These are the single electron states created by the lateral quantization; the splittings range from 2 to 8 meV. The structure is not expected to be regular (as seen in the single-dot observations), as it is a complicated superposition of two spectra. The structure is repeatable, and weakens considerably by ~ 30 K though it persists to higher temperature. The splittings of ~ 8 meV are in rough agreement with the physical size (8 meV implies a diameter of $\sim 0.3 \mu\text{m}$).

Figure 5 is current-voltage data taken at 4.2 K from a device which is $\sim 0.18 \mu\text{m}$ in diameter. At least seven resonances are evident. We are in the early stages of data interpretation.

The intriguing observation here is that relatively large

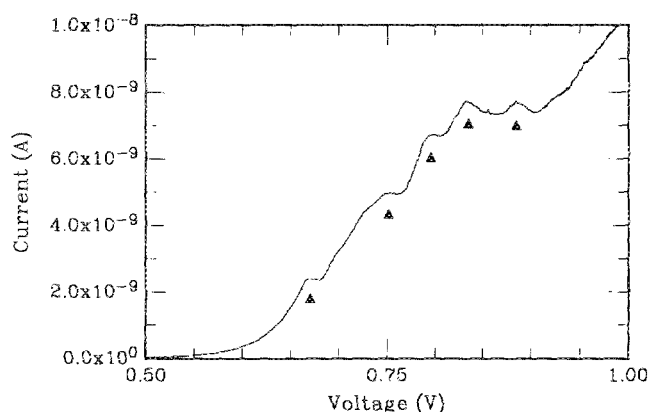


FIG. 3. I - V characteristics of a single quantum dot nanostructure at 1.0 K showing resonant tunneling through the discrete states of the $n = 2$ quantum well resonance. \blacktriangle indicates the voltage positions of the discrete states for the $T = 1.0$ K curve.

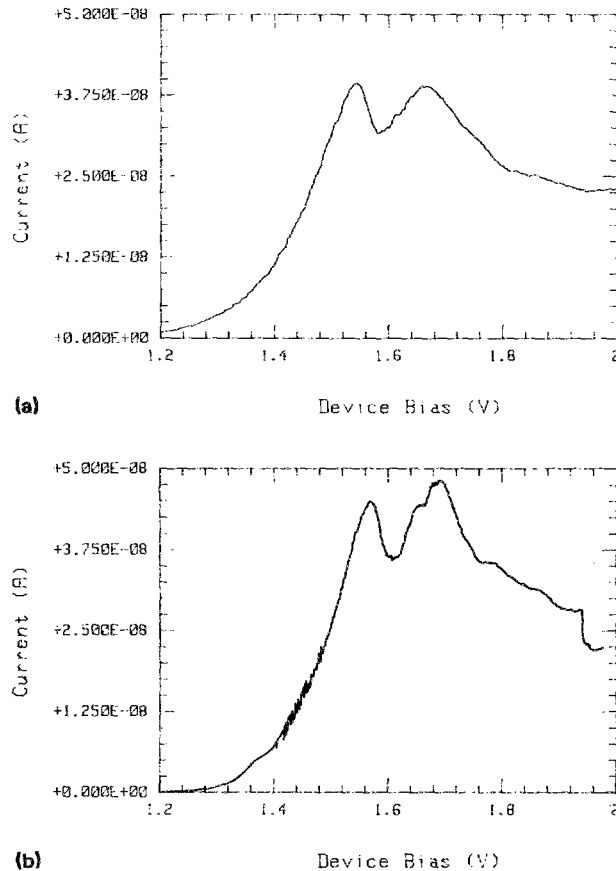


FIG. 4. (a) I - V characteristics of a 60-Å $\text{Al}_{0.4}\text{Ga}_{0.6}\text{As}$ barrier/50-Å GaAs quantum well/35-Å $\text{Al}_{0.4}\text{Ga}_{0.6}\text{As}$ barrier/80-Å GaAs quantum well/60-Å $\text{Al}_{0.4}\text{Ga}_{0.6}\text{As}$ barrier structure (double dot), fabricated with a diameter of $\sim 0.25 \mu\text{m}$. $T = 100$ K. (b) I - V characteristics of the same device at $T = 4.2$ K.

quantum dots in this material exhibit lateral quantization. Previous measurements on single-dot structures fabricated from double-barrier/single quantum well material and also of $\sim 0.25\text{-}\mu\text{m}$ diameter did not exhibit structure. However, the current density of the resonance in this double quantum well structure is almost 1000 times less than the single quantum well structure. A tentative hypothesis is that the large effective barrier widths in the double-dot structure both decrease the tunneling component, which is strictly longitudinal (which could mask the transverse quantization effects),

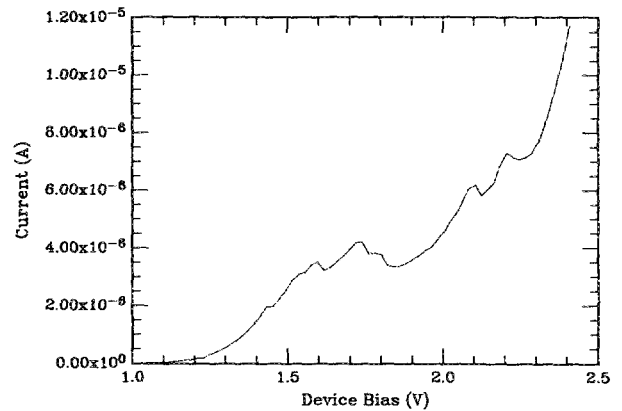


FIG. 5. I - V characteristics of a double-dot diode fabricated with a diameter of $\sim 0.18 \mu\text{m}$. $T = 4.2$ K.

and increases the mixing of the transverse and longitudinal wave functions. Also, the coupling between the two dots may enhance the mixing of wave functions.

V. CONCLUSIONS

We have described the fabrication processes for producing quantum dot diodes. Transport measurements through single- and double-dot diodes have been presented which demonstrate the discrete electronic states due to the zero-dimensional nature of the dots.

ACKNOWLEDGMENTS

The authors gratefully acknowledge the technical contributions of Elaine Pijan, Pam Stickney, Bob Aldert, Randy Thomason, and Doug Schultz. We would also like to thank Bob Bate, Bill Frensley, Chia-Hung Yang, Jim Luscombe, and Alan Seabaugh for enlightening discussions. This work was sponsored in part by AFWAL, ARO, and ONR.

¹R. T. Bate, *Superlattices Microstructures* 2, 9 (1986).

²J. N. Randall, M. A. Reed, T. M. Moore, R. J. Matyi, and J. W. Lee, *J. Vac. Sci. Technol. B* 6, 302 (1988).

³L. L. Chang, L. Esaki, and R. Tsu, *Appl. Phys. Lett.* 24, 593 (1974).

⁴M. A. Reed, J. N. Randall, R. J. Aggarwal, R. J. Matyi, T. M. Moore, and A. E. Wetsel, *Phys. Rev. Lett.* 60, 535 (1988).

⁵S. Y. Chou, E. Wolak, and J. S. Harris, *Appl. Phys. Lett.* 52, 657 (1988).

⁶A. E. Wetsel, M. A. Reed, J. N. Randall, R. J. Aggarwal, R. J. Matyi, and T. M. Moore (unpublished).